

**PCT**WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau

## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification <sup>6</sup> : <b>A61K 6/00, 7/06</b>		<b>A1</b>	(11) International Publication Number: <b>WO 00/13653</b>
			(43) International Publication Date: 16 March 2000 (16.03.00)
(21) International Application Number: <b>PCT/US99/13043</b>			(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).
(22) International Filing Date: <b>9 June 1999 (09.06.99)</b>			
(30) Priority Data: <b>09/149,964 9 September 1998 (09.09.98) US</b>			
(71) Applicant: <b>ARCHIMICA (FLORIDA), INC. [US/US]; P.O. Box 1466, Gainesville, FL 32602-1466 (US).</b>			
(72) Inventors: <b>LEGROW, Gary, E.; 13707 S.W. 1st Lane, Newberry, FL 32669 (US). TERRY, W., Leonard, Jr.; 4830 N.W. 43rd Street, I-131, Gainesville, FL 32606 (US).</b>			
(74) Agent: <b>CLEMENT, Alan, B.; Hedman, Gibson &amp; Costigan, P.C., 1185 Avenue of the Americas, New York, NY 10036 (US).</b>			Published <i>With international search report.</i>
(54) Title: <b>ORGANOSILICONE COMPOSITIONS FOR PERSONAL CARE</b>			
(57) Abstract <p>The present invention relates to compositions comprising non-toxic and safe for human contact and ingestion volatile or non-volatile silicone-aliphatic hydrocarbon hybrid fluids and a non-volatile silicone-aliphatic hydrocarbon hybrid wax which have the physical consistency of a gel which are of use in a wide variety of personal care applications and when applied to the human skin components within these compositions adsorb quickly into the upper layers of the stratum corneum, while other components form non-tacky occlusive films on the outer surface of the stratum corneum.</p>			

*FOR THE PURPOSES OF INFORMATION ONLY*

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece	ML	Mali	TR	Turkey
BG	Bulgaria	HU	Hungary	MN	Mongolia	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MR	Mauritania	UA	Ukraine
BR	Brazil	IL	Israel	MW	Malawi	UG	Uganda
BY	Belarus	IS	Iceland	MX	Mexico	US	United States of America
CA	Canada	IT	Italy	NE	Niger	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NL	Netherlands	VN	Viet Nam
CG	Congo	KE	Kenya	NO	Norway	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NZ	New Zealand	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	PL	Poland		
CM	Cameroon	KR	Republic of Korea	PT	Portugal		
CN	China	KZ	Kazakhstan	RO	Romania		
CU	Cuba	LC	Saint Lucia	RU	Russian Federation		
CZ	Czech Republic	LI	Liechtenstein	SD	Sudan		
DE	Germany	LK	Sri Lanka	SE	Sweden		
DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

WO 00/13653

PCT/US99/13043

---

**ORGANOSILICONE COMPOSITIONS FOR PERSONAL CARE**

---

**FIELD OF THE INVENTION**

The present invention relates to compositions for use in personal care products. More particularly, the present invention relates to compositions for use in personal care products which have the physical form of a gel. Most particularly, the present invention relates to compositions for use in personal care products which when applied to the human skin, have component(s) which adsorb quickly into the upper layers of the stratum corneum, and have component(s) which form non-tacky occlusive films on the outer surface of the stratum corneum. Significantly, all of the components employed in the use of the compositions of the present invention are safe for human contact and ingestion.

**BACKGROUND OF THE PRESENT INVENTION**

It is well known in the art that an organosilicone gum can be used to thicken a cosmetic formulation containing organic components and organosilicone components such as dimethylcyclodimethylsiloxanes and low molecular weight polydimethylsiloxanes. More recently, new silicone technologies have been found to thicken cosmetic formulations including silicone copolyols and their derivatives (United States Patent No. 5,623,017 and 5,336,497), and lightly crosslinked silicone hydrosilylation addition compositions (United States Patent Nos. 5,679,734 and 5,306,797). Thickening of cosmetic formulations is typically performed to minimize the product running on or from the surface to which it is applied.

It is also well known in the art that polydimethylsiloxanes, including gums and elastomeric materials, have a larger free volume than typical organic materials, thereby allowing water and other vapors to pass through them as readily as they pass through air. Consequently formulations containing silicone gums and elastomers generally do not form occlusive barriers on the skin. It is also well known in the art that polydimethylsiloxanes, including gums and elastomers, are not compatible with most organic materials used in personal care

WO 00/13653

PCT/US99/13043

formulations, thereby making the development of homogeneous formulations with these ingredients difficult.

New information has recently been published about toxicity characteristics of octamethylcyclotetrasiloxane which is a common silicone component of many cosmetic formulations. See, SEHSC Report entitled "D4 (Octamethylcyclotetrasiloxane) Scientific Paper for Use by Member Companies" dated July 20, 1998. This information would suggest that non-toxic alternatives to the use of this material would be desirable, and preferred.

The inclusion of a volatile alkylmethylsiloxane in a formulation with octamethylcyclotetrasiloxane and a polydimethylsiloxane gum has recently been described in United States Patent No. 5,759,529 for the purpose of enhancing the rate of adsorption, and improving general aesthetic characteristics.

It would therefore represent a significant advance in the state of the art if a siloxane formulation could be provided which employed nontoxic silicone components in combination with a gelling agent which had improved compatibility characteristics with other cosmetic ingredient constituents, and which had substantially equivalent sensory characteristics to current siloxane cosmetic formulations.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a chart showing a comparison of sensory profiles of volatile silicones as described in Example 1 of the present application.

Figure 2 is a chart showing a comparison of sensory profiles of gelled silicones as described in Example 2 of the present application.

#### SUMMARY OF THE INVENTION

The present invention is directed to improvements in thickened cosmetic formulations which employ a thickening component which does not suffer from the incompatibility problems associated with polydimethylsiloxane gums and elastomers as discussed hereinabove. Further, the present invention is directed to improvements in thickened cosmetic formulations which eliminate potentially toxic ingredients, such as

WO 00/13653

PCT/US99/13043

5 octamethylcyclotetra-siloxane, which are effective in providing an occlusive film on the outer layer of the skin after application, and which maintains or improves upon the aesthetic characteristics of the prior art formulations, during and after application.

10 To this end the present invention provides a composition having the physical consistency of a gel wherein said composition comprises (a) a volatile or non-volatile silicone-hydrocarbon hybrid fluid, and (b) a gelling agent comprising a non-volatile occlusive silicone-aliphatic hydrocarbon hybrid wax. Aliphatic hydrocarbons are well known to be occlusive, especially as their chain length and molecular weight increase. Exemplary of such materials is petrolatum. On the other hand, silicones are generally not occlusive. Hybridization of these two types of  
15 constituents can produce an infinite array of silicone-aliphatic hydrocarbon hybrid materials ranging from volatile non-occlusive fluids to non-volatile high softening point occlusive waxes.

20 Typical of the volatile non-occlusive silicone-aliphatic hydrocarbon hybrid fluids useful in the practice of the present invention is 3-n-hexyl-1,1,1,3,5,5,5-heptamethyltrisiloxane. This chemical is known to have a volatility similar to that of dimethylcyclsiloxanes such as octamethylcyclotetrasiloxane and decamethylcyclpentasiloxane. Surprisingly, the sensory characteristics of this chemical are  
25 virtually identical to those of octamethylcyclotetrasiloxane.

. Typical of the non-volatile occlusive high softening point waxes useful in the practice of the present invention is the wax Poly (n-C24-28 Alkylmethylsiloxane)dimethylsiloxane, referred to as C24-28 Alkyl Dimethicone by INCI nomenclature as  
30 used in the cosmetic industry. This wax has a softening point of 65 degrees Centigrade and has a hard consistency at room temperature such that it cannot be spread onto the skin to form a continuous film under ambient conditions.

35 In a preferred embodiment a mixture of 10% w/w C24-28 Alkyl Dimethicone wax and 90% w/w 3-n-hexylheptamethyltrisiloxane is prepared and heated above the softening point of the wax to

WO 00/13653

PCT/US99/13043

produce a clear solution. On cooling of the solution to ambient temperature a gel is formed. In spite of the high softening point of the wax alone, this gel has silicone-like aesthetic characteristics and can be readily spread on the skin. As the volatile organosilicone evaporates and adsorbs into the upper layers of the stratum corneum a film of the C24-28 Alkyl Dimethicone wax is left on the surface of the stratum corneum which also has silicone-like aesthetic characteristics in spite of its 70% hydrocarbon content.

It has been found that a range of Poly(alkylmethylsiloxane)dimethylsiloxane copolymers with a hydrocarbon content of at least 60%, an average molecular weight of about 2000 or above, and a softening point of about 50°C or above will gel volatile fluids of many kinds including, dimethylsilicones, silicone-aliphatic hydrocarbon hybrids, and hydrocarbons at as low as 5% w/w.

Based upon this finding, it is now possible to prepare cosmetic formulations which do not run on the skin, which possess no dimethylcyclosiloxanes, which have dimethylsilicone-like aesthetically pleasing characteristics during application, and which form occlusive films which again are aesthetically pleasing in accordance with the present invention. Furthermore, the difficulty of using dimethylsilicone gums to thicken cosmetic formulations can be avoided. Other cosmetic ingredients, both organic and organosilicone, both inactive and active, can be readily formulated on a uniform basis into these gels.

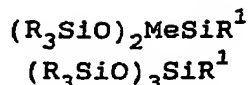
#### DETAILED DESCRIPTION OF THE INVENTION

The present invention provides a composition having the physical consistency of a gel wherein said composition comprises (a) a volatile or non-volatile silicone-hydrocarbon hybrid fluid, and (b) a gelling agent comprising a non-volatile occlusive silicone-aliphatic hydrocarbon hybrid wax.

Volatile non-occlusive silicone-aliphatic hydrocarbon hybrid fluids useful in the practice of the present invention are any of those known to those skilled in the art. Preferred are fluids having the general formulae:

WO 00/13653

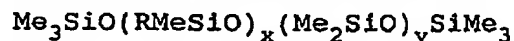
PCT/US99/13043



wherein each R is independently the same or different monovalent straight or branched chain alkyl group having from 1 to about 3 carbon atoms, and R<sup>1</sup> is a monovalent straight or branched chain alkyl group having from 6 to about 18 carbon atoms or is phenyl. These are prepared by methods known to those skilled in the art.

Preferred volatile or non-volatile silicone-hydrocarbon hybrid fluids of the present invention are those with sensory characteristics similar to those of octamethylcyclotetrasiloxane, which include 3-n-hexyl-1,1,1,3,5,5,5-heptamethyltrisiloxane, 3-n-octyl-1,1,1,3,5,5,5-heptamethyltrisiloxane, n-hexyltris(trimethylsiloxy)silane, phenyltris(trimethylsiloxy)silane and n-octyltris(trimethylsiloxy)silane. Other useful fluids are those such as, but not limited to 3-n-dodecyl-1,1,1,3,5,5,5-heptamethyltrisiloxane and 3-n-octadecyl-1,1,1,3,5,5,5-heptamethyltrisiloxane. Of course mixtures of any of the fluids may be employed in the practice of the present invention. In preferred embodiments, the fluids employed are at least 99% pure.

The second component of the compositions of the present invention comprises non-volatile occlusive silicone-aliphatic hydrocarbon hybrid waxes. These can be prepared according to methods known to those skilled in the art. Particularly useful in the practice of the present invention are poly(n-alkylmethylsiloxane)dimethylsiloxanes with n-alkyl substitution of an average of 22 carbon atom or above, with an average of more than 2 alkyl groups per molecule, with hydrocarbon contents of at least 60% and with average molecular weights of at least 2000 or more. Examples of waxes preferred for use in accordance with the present invention are random copolymers of the following formula:



wherein R is an aliphatic hydrocarbon substituent with at least an average of a 22 carbon atom chain, x is at least an average of more than 2, and y is at least 1. Specific examples include R = C<sub>20</sub>-C<sub>24</sub> and C<sub>24</sub>-C<sub>28</sub>, x=5 and y=3.

The above-described volatile fluids, alone or in any

WO 00/13653

PCT/US99/13043

combination, can be gelled with as little as 5% w/w, and up to about 40% or more w/w of any of the above poly(n-alkylmethylsiloxane)dimethylsiloxane waxes. In some embodiments from about 10% to about 20% w/w of wax is employed.

5           The gels can also be formulated into cosmetic formulation with the addition of other cosmetic ingredients known to those skilled in the art including non-volatile silicone-aliphatic hydrocarbon hybrid fluids, and organic materials such as long chain aliphatic hydrocarbons and esters. Active  
10 ingredients such as stearoxytrimethylsilane can be readily formulated into these mixtures. Other cosmetic additives including perfumes, antiperspirant, a humectant, an insect repellent, an odorant, a deodorant, an emollient, an antiseptic, a sunscreen, a cleansing agent, a suitable pharmaceutical, a  
15 pigment, a biocide and mixtures of any of the foregoing may also be added.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

The following examples illustrate the present invention. They are not to be construed to limit the scope of the  
20 appended claims in any matter whatsoever.

#### EXAMPLE 1

Following the protocol of ASTM method E 1490-92 entitled "Sensory Evaluation of Materials and Products," a Sensory Panel composed of 24 volunteers was established and  
25 trained using reference anchors. Sensory profiles of commercially available silicone products used in the cosmetic industry were determined by this trained panel and shown to match (within experimental error) the published profiles of these products.

Sensory profiles were determined for the following  
30 compounds: octamethylcyclotetrasiloxane (D4), 3-n-hexyl-1,1,1,3,5,5,5-heptamethyltrisiloxane (41M10), 3-n-octyl-1,1,1,3,5,5,5-heptamethyltrisiloxane (41M15), and n-octyltris-(trimethylsiloxy)silane (31M60). The averaged values for each sensory parameter obtained by the sensory panel are shown and  
35 plotted on Chart 1. It may be concluded from these data that the sensory profiles of these materials are very similar.



WO 00/13653

PCT/US99/13043

## EXAMPLE 2

Each of the volatile fluids identified in EXAMPLE 1 was gelled using  $C_{24}$ - $C_{28}$  Alkyl Dimethicone wax. This was accomplished in each case by adding 5 g. of  $C_{24}$ - $C_{28}$  Alkyl Dimethicone wax to 45 g. of each of the volatile fluids D4, 41M10, 41M15, and 31M60 and then heating the mixtures to approximately 70°C. Clear solutions of the molten wax in the volatile fluids were then allowed to cool back to room temperature. In each case a rigid, non-flowing gel was formed. The sensory characteristics of each of these gels was then determined by the Sensory Panel. The averaged values for each parameter obtained by the sensory panel are shown and plotted on Chart 2.

## EXAMPLE 3

In the same manner as described in EXAMPLE 2, 50 g. molten solutions of  $C_{24}$ - $C_{28}$  Alkyl Dimethicone wax (41M80) in the volatile silicone-aliphatic hydrocarbon fluid 41M10 were prepared at concentrations of 2.5%, 3.5%, 5%, 10% and 20% wax. On cooling to ambient temperature the volatile fluid was completely gelled in all of the samples with 5% wax or more. The samples with 2.5% and 3.5% wax were not fully gelled.

## EXAMPLE 4

In the same manner as described in EXAMPLE 2, molten 50 g. solutions of  $C_{20}$ - $C_{24}$  Alkyl Dimethicone and  $C_{24}$ - $C_{28}$  Alkyl Dimethicone waxes in the volatile fluid 41M10 were prepared at concentrations of 5% and 10% wax. On cooling to ambient temperature the volatile fluid was completely gelled in all 4 samples.

## EXAMPLE 5

In the same manner as described in EXAMPLE 2, a molten solution of 5 g. of  $C_{24}$ - $C_{28}$  Alkyl Methicone wax, (3-n-( $C_{24}$ - $C_{28}$ ) Alkyl-1,1,1,3,5,5,5-heptamethyltrisiloxane), in 45 g. of 41M10 fluid was prepared at 70°C. On cooling to room temperature, this solution showed no indication of gellation.

## EXAMPLE 6

In the same manner as described in EXAMPLE 2, a molten solution of 5 g. of  $C_{24}$ - $C_{28}$  Alkyl Dimethicone (41M80) in 45 g. of

WO 00/13653

PCT/US99/13043

50 cs. mineral oil was prepared at 70°C. On cooling to ambient temperature, the mineral oil was completely gelled to a non-flowing substance.

## EXAMPLE 7

5 In the same manner as described in EXAMPLE 2, the following solutions were prepared at approximately 70°C and then allowed to cool to ambient temperature. In all cases the mixtures were fully gelled.

10 (a) 5 g. 41M80 wax, 5 g. C<sub>24</sub>-C<sub>28</sub> Alkylheptamethyltrisiloxane wax, and 40 g. 41M10 volatile fluid.

(b) 5 g. 41M80 wax, 5 g. 3-n-Stearylheptamethyltrisiloxane non-volatile fluid, and 40 g. 41M10 volatile fluid.

(c) 5 g. 41M80 wax, 5 g. of Stearoxytrimethylsilane, and 40 g. 41M10 volatile fluid.

15 (d) 5 g. 41M80 wax, and 45 g. Phenyltris(trimethylsiloxy)-silane.

(e) 1 g. Dow Corning AMS-C30 wax and 19 g. 41M10 volatile fluid.

20 Variations of the present invention will suggest themselves to those skilled in the art in light of the above detailed description. For example, a mixture of any combination of volatile silicones described above could be gelled to generate a gel with a sensory profile unlike any of the profiles described above. Other volatile silicones including other cyclosiloxanes,  
25 such as decamethylcyclopentasiloxane, or phenyl containing siloxanes may be gelled in the same manner as described above.

The above-referenced patents and publications are hereby incorporated by reference in their entirety.

WO 00/13653

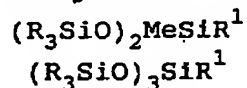
PCT/US99/13043

CLAIMS

1. A composition having the physical consistency of a gel wherein said composition comprises (a) a volatile or non-volatile silicone-hydrocarbon hybrid fluid, and (b) a gelling agent comprising a non-volatile occlusive silicone-aliphatic hydrocarbon hybrid wax.

2. A composition as defined in Claim 1 wherein said components (a) and (b) are known either directly or by inference with other related materials to be safe for human contact or ingestion.

3. A composition as defined in Claim 1 wherein said volatile or non-volatile silicone-hydrocarbon hybrid fluid (a) comprises a compound of the general formulae:



wherein Me is methyl, each R is independently the same or different monovalent straight or branched chain alkyl group having from 1 to about 3 carbon atoms, and  $R^1$  is a monovalent straight or branched chain alkyl group having from 6 to about 18 carbon atoms or phenyl.

4. A composition as defined in Claim 3 wherein said volatile silicone-hydrocarbon hybrid fluid (a) is selected from the group consisting of 3-n-hexyl-1,1,1,3,5,5,5-heptamethyltrisiloxane, 3-n-octyl-1,1,1,3,5,5,5-heptamethyltrisiloxane, n-hexyltris(trimethylsiloxy)silane, phenyltris(trimethylsiloxy)silane, n-octyltris(trimethylsiloxy)silane and mixtures of any of the foregoing.

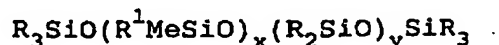
5. A composition as defined in Claim 3 wherein said volatile silicone-hydrocarbon hybrid fluid (a) is at least about 99% pure.

6. A composition as defined in Claim 3 wherein said non-volatile silicone-hydrocarbon hybrid fluid is selected from the group consisting of 3-n-dodecyl-1,1,1,3,5,5,5-heptamethyltrisiloxane, 3-n-octadecyl-1,1,1,3,5,5,5-heptamethyltrisiloxane and mixtures thereof.

WO 00/13653

PCT/US99/13043

7. A composition as defined in Claim 1 wherein said non-volatile silicone-hydrocarbon hybrid wax (b) comprises a polymer of the general formula



5 wherein Me is methyl, each R is independently the same or different monovalent straight or branched chain alkyl group having from 1 to about 3 carbon atoms,  $R^1$  is a monovalent straight or branched chain alkyl group having from an average of about 22 to an average of about 28 carbon atoms, x is an average  
10 of more than 2, y is 1 or more, and the molecular weight of the polymer is about 2000 or more.

8. A composition as defined in Claim 1 wherein said non-volatile silicone-hydrocarbon hybrid wax (b) comprises a polymer of the general formula



5 wherein Me is methyl each R is independently the same or different monovalent straight or branched chain alkyl group having from 1 to about 3 carbon atoms,  $R^1$  is a monovalent straight or branched chain alkyl group having an average of about 22 to or more, x is an average of more than 2, and the molecular  
10 weight of the polymer is about 2000 or more.

9. A composition as defined in Claim 7 wherein said non-volatile silicone-hydrocarbon hybrid wax is selected from  $Me_3SiO(C_{20}-C_{24}alkylMeSiO)_5(Me_2SiO)_3SiMe_3$ , and  $Me_3SiO(C_{24}-C_{28}AlkylMeSiO)_5(Me_2SiO)_3SiMe_3$  waxes and mixtures of the foregoing.

10. A composition as defined in Claim 1 wherein said non-volatile silicone-hydrocarbon hybrid wax is present in an amount ranging from about 5% to about 40% (w/w).

11. A composition as defined in Claim 1 wherein said non-volatile silicone-hydrocarbon hybrid wax is present in an amount ranging from about 10% to about 20% (w/w).

12. A composition as defined in Claim 1 further comprising cosmetic additives.

WO 00/13653

PCT/US99/13043

13. A composition as defined in Claim 12 wherein said cosmetic additives comprises stearoxytrimethylsilane.

14. A method of treating dry skin comprising delivering to the stratum corneum with an effective amount of a composition as defined in Claim 1.

WO 00/13653

PCT/US99/13043

1/2

## SENSORY CHARACTERISTICS OF VOLATILE SILICONES

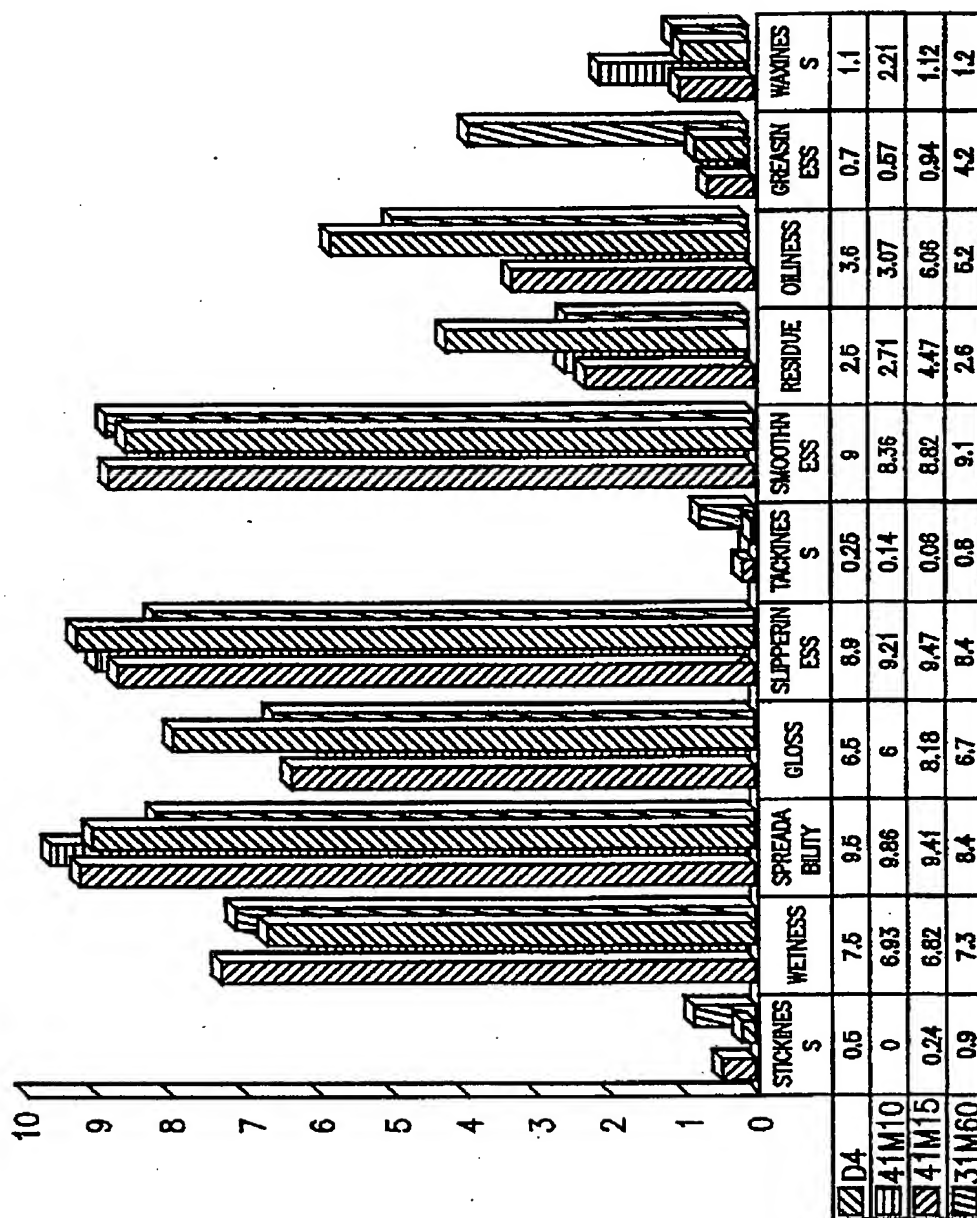


FIG. 1

SUBSTITUTE SHEET (RULE 26)

WO 00/13653

PCT/US99/13043

2/2

SENSORY PROFILES OF 10% 41M80 IN VOLATILE SILICONES

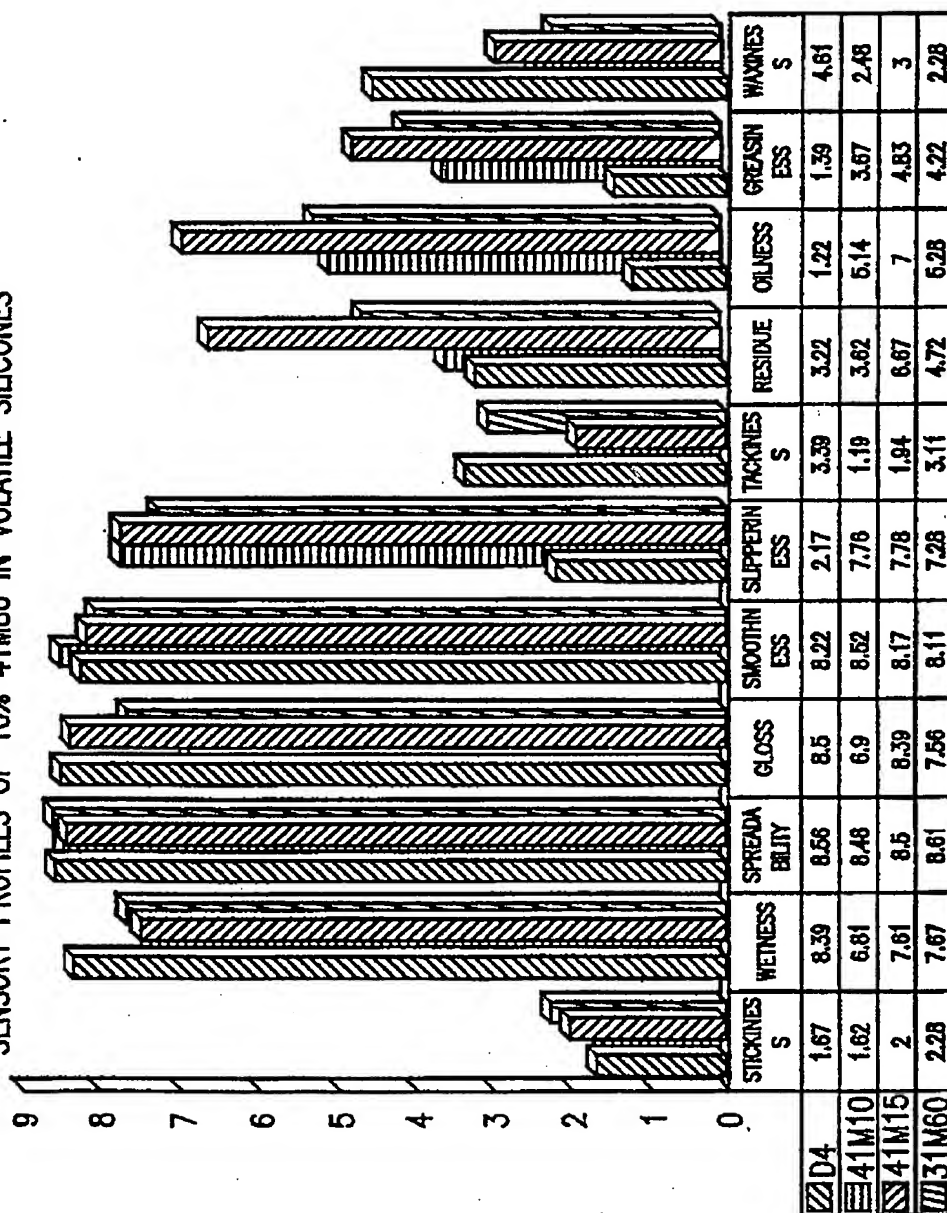


FIG. 2

SUBSTITUTE SHEET (RULE 26)

## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US99/13043

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : A61K 6/00; 7/06

US CL : 424/400, 401, 70.11, 70.12; 514/944

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 424/400, 401, 70.11, 70.12; 514/944

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
WEST

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5,225,188 A (ABRUTYN et al) 06 July 1993, see entire document.	8
Y	US 5,413,781 A (GIWA-AGBOMEIRELE et al) 09 May 1995, see entire document.	1-7 and 9-14
Y	US 5,492,691 A (BAHR et al) 20 February 1996, see entire document.	1-7 and 9-14

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

## \* Special categories of cited documents:

\*A\* document defining the general state of the art which is not considered to be of particular relevance

\*B\* earlier document published on or after the international filing date

\*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

\*O\* document referring to an oral disclosure, use, exhibition or other means

\*P\* document published prior to the international filing date but later than the priority date claimed

\*T\*

later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

\*X\*

document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

\*Y\*

document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

\*A\*

document member of the same patent family

Date of the actual completion of the international search

13 SEPTEMBER 1999

Date of mailing of the international search report

7 October 1999

Name and mailing address of the ISA/US  
Commissioner of Patents and Trademarks  
Box PCT  
Washington, D.C. 20231

Facsimile No. (703) 305-3230

Authorized officer

LAKSHMI S. CHANNAVAJALA

Telephone No. (703) 308-0196